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POLYACETAL RESIN COMPOSITE  
[Poriasetaaru jushi soseibutsu]

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## Specification

### Title of the Invention

Polyacetal Resin Composite

### Claims

1 Polyacetal resin composite so characterized that in a polyacetal resin composite comprised by distributing 1 to 100 parts by weight granular inorganic filler material with an average grain size of 100  $\mu\text{m}$  or less in 100 parts by weight polyacetal resin, said granular inorganic filler material is surface-treated ahead of time by 0.1 to 20 wt% (of said granular inorganic filler material) of one or more types of surface treating agent or adhering agent selected from fatty acids with 12 or more carbon atoms and their metallic salts.

2 Polyacetal resin composite described in Claim 1 wherein the main structural component of granular inorganic filler material is an oxide, hydroxide, carbonate, silicate, or sulfate of a Periodic Table Group II or Group III metal, or silicon dioxide.

3 Polyacetal resin composite described in Claim 1 wherein the granular inorganic filler material is calcium carbonate or talc.

4 Polyacetal resin composite described in any of Claims 1 to 3 wherein the surface treating agent or adhering agent is a saturated fatty acid with 12 to 30 carbon atoms.

### Detailed Description of the Invention

#### (Industrial Field of Application)

This invention pertains to a polyacetal resin composite that

maintains a good balance between the intrinsic strength of polyacetal resins and superior properties such as toughness, and has minimal glossiness.

(Prior Art and Its Problems)

As is well known, polyacetal resins are used in an extremely wide range of fields as typical engineering resins having physical characteristics such as mechanical properties and electrical properties or chemical characteristics such as chemical resistance and heat resistance. As fields using polyacetal resins have broadened, however, greater specificity is demanded in their properties as materials.

For purposes such as minimizing irritation of eyes by reflecting light, producing a feeling of high quality, or preventing mechanical malfunction due to reflected light, one of these properties that is occasionally demanded in fields such as automotive interior products or optical machinery is little gloss; that is, little reflection of light. To respond to this demand, methods for adding an inorganic filler such as calcium carbonate or talc to polyacetal resin is known in prior art.

Although these methods offered by prior art obtain polyacetal resins having desirable properties in terms of reduced gloss, in return for this, they have the drawbacks that resins have reduced mechanical characteristics, especially extension and toughness, and are easily damaged when subjected to stress during working or assembly of molded products or impact such as dropping during handling of molded products, and there is a demand to correct these drawbacks.

(Means of Solving the Problems)

As a result of extensive research to obtain polyacetal resin

composites that correct the drawbacks described above, the present inventors discovered that optimum results were obtained by distributing granular inorganic filler material that had been surface-treated ahead of time by a specific treating agent, and were able to perfect the present invention.

That is, this invention pertains to a polyacetal resin composite so characterized that in a polyacetal resin composite comprised by distributing 1 to 100 parts by weight granular inorganic filler material with an average grain size of 100  $\mu\text{m}$  or less in 100 parts by weight polyacetal resin, said granular inorganic filler material is surface-treated ahead of time by 0.1 to 20 wt% (of said granular inorganic filler material) of one or more types of surface treating agent or adhering agent selected from fatty acids with 12 or more carbon atoms and their metallic salts.

For the polyacetal resin used in this invention, any polyacetal resins can be used, such as polyoxydimethylene homopolymer or polyacetal copolymers or terpolymers having a main chain comprised mostly of an oxymethylene chain. In addition, there are no special restrictions on factors such as degree of polymerization.

Granular inorganic filler material that has been surface-treated by a specific treating agent is distributed in such a polyacetal resin.

The granular inorganic filler material used here tends to damage the external appearance of molded products by producing flow marks or forming irregularities as grain size increases, and this tendency becomes marked at or above a certain grain size. Therefore, in this invention, granular material with an average grain size of 100  $\mu\text{m}$  or

less is used as the inorganic filler material. Preferably, the average grain size is 0.01 to 50  $\mu\text{m}$ , and especially preferably, 0.05 to 10  $\mu\text{m}$ .

In addition, there are no special restrictions on the type of granular inorganic filler material used in this invention, but considering efficient reduction of gloss, which is the purpose of this invention, as well as various properties, the main structural component is preferably an oxide, hydroxide, carbonate, silicate, or sulfate of a Periodic Table Group II or Group III metal, or silicon dioxide. Examples of these include calcium oxide, calcium carbonate, magnesium carbonate, calcium sulfate, barium sulfate, talc, clay, kaolin, bentonite, dolomite, and silica. Using calcium carbonate or talc is especially preferred.

In this invention, granular inorganic filler material such as described above must be surface-treated ahead of time by a specific organic compound. The surface treating agent or adhering agent used in this surface treatment is selected from fatty acids with 12 or more carbon atoms and their metallic salts.

Examples of such fatty acids include lauric acid, trimellitic acid, myristic acid, pentadecanoic acid, palmitic acid, heptadecanoic acid, stearic acid, nonadecanoic acid, arachidic acid, behenic acid, lignoceric acid, cerotic acid, heptacosane acid, montanic acid, melissic acid, lacseric [as transliterated] acid, oleic acid, elaidic acid, linoleic acid, linolenic acid, arachidonic acid, setoleic [as transliterated] acid, and erucic acid.

Metallic salts of these fatty acids include calcium salts, magnesium salts, zinc salts, barium salts, sodium salts, and aluminum

salts.

Of the above, fatty acids are especially ideal in this invention. Of these, saturated fatty acids with 12 to 30 carbon atoms are extremely effective.

The method for treating inorganic filler material by such surface treating agents or adhering agents in this invention is not subject to special restrictions, and any known method can be used. For example, either of the following methods can be used: the method of agitating and blending inorganic filler material and treating agents at or above the melting point of the treating agents; or the method of dissolving surface treating agents in a solvent, agitating and blending this with inorganic filler material, then burning off and eliminating the solvent.

The granular inorganic filler material used in this invention uses compounds such as described above, and is surface-treated such that the weight of surface treating agents or adhering agents is 0.1 to 20 wt% of inorganic filler material.

At weight less than 0.1 wt%, surface treating agents do not obtain the improvement in mechanical strength, especially extension and toughness, that is the purpose of this invention, while weight greater than 20 wt% leads to reduced molded product strength and impaired composite stability. The preferred weight of surface treating agents is 0.5 to 10 wt%, and more preferably 1 to 7 wt%, of inorganic filler material.

The amount of granular inorganic filler material surface-treated in this way added to polyacetal resin in this invention is 1 to 100 parts by weight per 100 parts by weight polyacetal resin. At an amount less

than this, adequate effect reducing gloss cannot be obtained, and an amount greater than this can lead to reduced physical properties or impaired heat stability. The preferred amount added is 5 to 70 parts by weight, and a range of 10 to 45 parts by weight is ideal.

Moreover, just distributing inorganic filler such as fatty acids in polyacetal resin without surface-treating granular inorganic filler material ahead of time does not obtain the desired properties. The desired properties are obtained for the first time by the method of this invention in which inorganic filler surface-treated ahead of time by such substances is distributed.

The composite of this invention can be reinforced in stability by also adding various types of stabilizers by prior art. Various additives by prior art can also be distributed to improve these properties as required by the intended application. To give examples, such additives include various types of coloring agents, lubricants, mold release agents, nucleating agents, surface active agents, heterogenous polymers, organic polymer improving agents, and fibrous, granular, or flake-form fillers such as inorganic, organic, or metallic fillers. One type or a mixture of two or more types of these can be used.

The composite of this invention is easily prepared by standard methods generally used to prepare resin composites by prior art. For example, any of the following methods can be used: the method of blending, then kneading and extruding components by an extruder to prepare pellets, then molding; the method of preparing pellets of different compositions first, blending set amounts of these pellets and supplying to molding, and obtaining the desired composite after molding;



or the method of charging a molding machine directly with one or more components.

#### (Working Examples)

Working examples of this invention will be explained below, but this invention is not limited to these working examples.

#### Working Examples 1 to 8 and Comparative Examples 1 to 5

Calcium carbonate or talc surface-treated ahead of time by the fatty acids or their metallic salts shown in Table 1 was added at the ratios shown in Table 1 to 100 parts by weight polyacetal resin ( trade name: "Duracon-M270" manufactured by Polyplastics Co., Ltd.), blended by a ribbon blender, then melted and kneaded using a biaxial extruder, and a pellet-shaped composite was obtained. Next, test pieces were molded from these pellets using an injection molder, then measured for various properties. For comparison, polyacetal resin with untreated calcium carbonate or talc added, polyacetal resin with calcium carbonate or talc and fatty acids simply added without surface-treating ahead of time by fatty acids, and polyacetal resin with no inorganic filler added were prepared and evaluated in the same way.

Results are shown in Table 1. Moreover, properties were measured as follows:

#### Gloss Measurement

Gloss was measured by a gloss meter (Model MGV4D manufactured by Suga Test Instruments) at an angle of incidence of 45° and angle of reflection of 45° using test pieces colored black by blending 0.5 wt% carbon black before molding.

#### Tensile Characteristics

Test pieces were left for 48 hours under conditions of 23°C temperature and 50% humidity, and measured in this same atmosphere as stipulated by ASTM D638 using a tensile tester (Tensilon manufactured by Orientek).

Table 1

	Additives					Physical Properties		
	Inorganic Filler Material			Other Additives		Gloss (%)	Tensile Characteristics	
	Type	[Amount Surface Treating Agent] <sup>2</sup>	Amount Added <sup>1</sup> (parts by weight)	Type	Amount Added <sup>1</sup> (parts by weight)		Strength (kgf/cm <sup>2</sup> )	Extension (%)
Working Example 1	calcium carbonate (average grain size: 4 μm)	[5 wt% stearic acid]	20	—	—	17.6	516	65
Working Example 2	calcium carbonate (average grain size: 4 μm)	[5 wt% stearic acid]	40	—	—	9.8	480	60
Working Example 3	calcium carbonate (average grain size: 4 μm)	[2 wt% stearic acid]	20	—	—	19.6	530	58
Working Example 4	calcium carbonate (average grain size: 4 μm)	[10 wt% stearic acid]	20	—	—	16.6	503	69
Working Example 5	calcium carbonate (average grain size: 4 μm)	[5 wt% stearic acid]	20	—	—	18.5	508	64
Working Example 6	calcium carbonate (average grain size: 4 μm)	[5 wt% calcium laurate]	20	—	—	17.8	512	47

Working Example 7	calcium carbonate (average grain size: 30 $\mu\text{m}$ )	[5 wt% stearic acid]	20	—	—	20.4	512	62
Comparative Example 1	—	[—]	—	—	—	92.6	610	48
Comparative Example 2	calcium carbonate (average grain size: 4 $\mu\text{m}$ )	[—]	20	—	—	17.6	505	20
Comparative Example 3	calcium carbonate (average grain size: 4 $\mu\text{m}$ )	[—]	19 <sup>*3</sup>	stearic acid	1 <sup>*3</sup>	17.8	508	23
Working Example 8	talc (average grain size: 3 $\mu\text{m}$ )	[5 wt% stearic acid]	20	—	—	13.1	526	45
Comparative Example 4	talc (average grain size: 3 $\mu\text{m}$ )	[—]	20	—	—	13.3	531	9
Comparative Example 5	talc (average grain size: 3 $\mu\text{m}$ )	[—]	19 <sup>*4</sup>	stearic acid	1 <sup>*4</sup>	13.5	522	12

\*1 amount added per 100 parts by weight polyacetal

\*2 concentration to inorganic filler material

\*3 same amount of inorganic filler material and treating agent as Working Example 1 just distributed

\*4 same amount of inorganic filler material and treating agent as Working Example 3 just distributed

(Effects of the Invention)

As is clear from the explanation and working examples described above, the polyacetal resin composite of this invention, in which granular inorganic filler material surface-treated ahead of time by specific fatty acids or their metallic salts has been distributed, can effectively reduce gloss on the surface of molded products, and has balanced physical properties without the reduction in extension and toughness found when conventional granular inorganic filler material is distributed. As a result, this invention is ideal for use in specialized applications such as automotive interior parts or optical machinery where reflected light from molded products is undesirable.